Cross-Section Standards for Neutron-Induced Gamma-Ray Production in the MeV Energy Range

R. O. Nelson*, N. Fotiades*, M. Devlin*, J. A. Becker†, P. E. Garrett†, and W. Younes†

*LANSCE-3, MS H855, Los Alamos National Laboratory, Los Alamos, NM 87545, USA
†Lawrence Livermore National Laboratory, Livermore, CA 94550, USA

Abstract. Gamma-ray cross-section standards for neutron-induced reactions are important in enabling the accurate determination of absolute cross sections from relative measurements of gamma-ray production. In our work we observed a need for improvement in these standards. In particular there are large discrepancies between evaluations of the $^{nat}Fe(n,n_1'\gamma)$ cross section for the 847-keV gamma ray. We have performed (1) absolute cross-section measurements, (2) measurements relative to the $^{nat}Cr(n,n_1'\gamma)$ 1434-keV gamma ray, and (3) comparisons using measured total and elastic scattering cross sections to refine our knowledge of the Fe cross section and the closely linked inelastic channel cross section for Fe. Calculation of integral tests of the cross section libraries may indicate that adjustment of the angular distributions of the neutron elastic and inelastic scattering may be needed.

INTRODUCTION

Measurements of γ-ray production cross sections provide data important for a variety of applications. Examples include determination of (n,2n) cross sections used in radiochemical diagnostics and determination of heating and damage in nuclear reactors and advanced nuclear systems. Iron is a common structural material and has also been proposed as providing a reasonable γ–ray cross-section standard. In work with GEANIE [1], the Germanium Array for Neutron-Induced Excitations at the WNR facility at LANSCE, to determine neutron-induced reaction cross sections we observed a need for better γ–ray cross-section standards, and in particular found that accuracy of the Fe 847-keV γ ray cross section was poor. Due to the near equivalence of the inelastic channel cross section and that for the 847-keV γ ray, these results also test the inelastic channel cross sections in the evaluated data libraries.

Measurement of neutron-induced γ rays from Fe date to 1935 with the experiments of Lea [2]. Use of the gamma-ray cross section to infer the inelastic channel cross section was mentioned as early as 1956 by Day [3]. Since then over 40 experiments have measured the (n,n'γ) cross section at various incident neutron energies. Despite these efforts agreement between the various measurements is generally poor, and various data evaluations reflect the uncertainties in the data by their differing values, as shown in Table 1.

TABLE 1. Evaluated library values for ^{56}Fe isotopic inelastic channel cross sections at E_n = 14.5 MeV.

Evaluation	Cross Section (mb)	% Difference from ENDF
ENDF/B-VI	681	0.0
BROND 2	610	-10.4
JEFF 3.0	724	6.3
JENDL 3.3	672	1.3

While neutron cross-section standards strive for 1% accuracy, the best available accuracy for γ-ray cross-section standards (other than for neutron capture) is typically 5% to 10%. In the case of the Fe(n,n'γ) reaction, two relatively recent evaluations [4,5] differ by 26%, while both claim accuracies of 5 to 10%. To address this problem we have performed (1) absolute cross-section measurements, (2) relative cross-section measurements using the Cr(n,n'γ) reaction, and (3) we use accurately measured neutron total cross-section and elastic-scattering data to further validate our cross-section results. We discuss discrepancies observed in

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integral measurements and possible solutions. Preliminary results for γ rays from Al, Si, and V also have been obtained.

We concentrate on the 14-MeV incident neutron energy region because angular distributions effects are smaller here, and because the effects of resonance structure and neutron-energy resolution do not have a strong influence on measurements at this energy.

EXPERIMENTS

The WNR facility at LANSCE and the GEANIE detector array are described by Lisowski *et al.* [6] and Nelson *et al.* [1], respectively. In brief, the LANSCE accelerator produces 800-MeV proton pulses with sub-nanosecond widths that are delivered to a water-cooled tungsten neutron production target. Spallation neutrons from the target travel a well-collimated 20.34-m flight path to the GEANIE sample.

In all measurements the γ-ray detector efficiency was determined *in situ* using calibrated γ-ray sources such as ^{152}Eu and mixed radionuclide sources.

The ^{nat}Fe(n,n$_1$'γ) 847-keV cross section at incident neutron energies above 11.4 MeV includes contributions from both ^{56}Fe(n,n'γ) and ^{57}Fe(n,2nγ) reactions. We present both the summed (^{56}Fe(n,n'γ) + ^{57}Fe(n,2nγ) photon cross-section result because it is useful as a standard cross section, and the pure ^{56}Fe(n,n'γ) cross section because it provides a measure of the inelastic channel cross section. We also present both natural and isotopic cross sections for convenience in comparisons.

Beta-decay of the ^{56}Mn product of the ^{56}Fe(n,p) reaction for $E_n > 3.0$ MeV, as well as contributions from the minor constituents ^{57}Fe(n,d) and ^{58}Fe(n,t) at higher energies, can pose a background problem for experiments that do not use pulsed beam techniques.

Absolute Cross-Section Measurements

Absolute γ-ray cross-section measurements have been made at our laboratory for many years. These measurements use a fission ionization chamber with both ^{238}U and ^{235}U foils to determine the incident neutron fluence [7]. The thickness of the U deposits and the distribution of material have been carefully measured using several techniques. The profile of the neutron beam spot is measured with x-ray image plates and used in MCNP simulations of the measurements to correct the detector acceptance. The contribution of neutron multiple scattering and reactions to the measured cross sections is performed using MCNP [8] calculations of the neutron flux in the sample and using a technique described in Nelson *et al.* [9]. MCNP calculations are also used to determine γ-ray attenuation in the samples.

The results of a 1994 measurement of the Fe(n,n'γ) 847-keV cross section are shown in Fig. 1 as the "Nelson 94" data points. Most of the published measurements are also shown for comparison. As expected from the near equivalence of the Fe inelastic channel cross section and the 847-keV partial γ–ray cross section, the shape of the ENDF inelastic excitation function and the shape of our data are very similar. This is not true for some of the previous measurements. We agree well with the excitation function shape of Dickens *et al.* [10] that was used in the ENDF evaluation; however our cross sections are about 20% larger than those of Dickens *et al.*

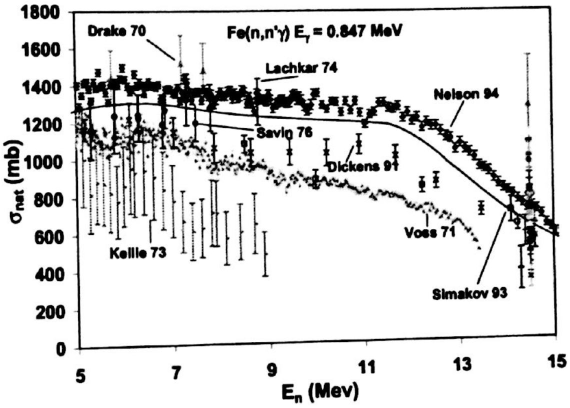

FIGURE 1. Absolute cross section measurements (most at 125 degrees) for the ^{nat}Fe(n,n'γ) $E_γ$ = 847 keV γ ray. Authors are indicated for some of the 34 experimental results plotted. The black curve is the ENDF/B-VI evaluation for the ^{56}Fe inelastic channel cross section multiplied by the 91.8% abundance of ^{56}Fe to convert to the "natural" cross section for comparison.

Relative Cross Section Measurements

Of the reactions proposed as γ–ray cross-section standards, the Cr(n,n'γ) 1434-keV cross section at 14.5 MeV appears to have the most consistent and accurate measurements. The evaluation by Simakov *et al.* [4] uses six measurements, all but one of which is consistent within uncertainties. The error in the evaluated cross section is 5%. There is one measurement on ^{52}Cr(n,n'γ) that is also in near agreement with the ^{nat}Cr measurements and has an uncertainty of 3.8%.

The measurements described here used relatively thin ^{nat}Fe and ^{nat}Cr plates. This reduces corrections for neutron multiple scattering and reactions as well as corrections for γ-ray attenuation in the samples. In this relative measurement, only the thickness of the samples, the relative efficiency for the γ rays, and the number of counts in the γ–ray peak are important. Corrections to the ratio include: γ–ray attenuation in the sample, multiple scattering and reactions in the sample, angular distribution effects, and internal conversion. All of these corrections were at the few percent level or less. A very minor correction was made for the ^{56}Fe(n,n$\alpha\gamma$)^{52}Cr reaction and the contribution of the ^{54}Fe(n,^{3}Heγ)^{52}Cr to the production of the 1434-keV γ ray was ignored.

Other γ rays that may provide suitable cross-section standards and for which we have preliminary results of relative measurements include: ^{27}Al, E_γ = 3004 keV; ^{nat}Si, E_γ = 1779 keV; and ^{nat}V, E_γ = 320 keV.

The ^{53}Cr(n,2nγ)^{52}Cr reaction contributes to the cross section of the 1434-keV γ ray for E_n > 9.5 MeV. The ^{53}Cr(n,2nγ)^{52}Cr reaction is included in the cross section value of 695 ± 35 mb from the evaluation of Simakov *et al.* [4] that we use to obtain the value for the Fe cross section from our relative measurement.

Because the slopes of the inelastic cross sections as a function of energy are very similar for Fe and Cr at incident-neutron energies near 14 MeV, the relative measurement is insensitive to small changes in the incident neutron energy. We used a neutron energy bin with a half width of 0.5 MeV centered on 14.5 MeV to obtain the cross section ratio. With a three-day measurement we obtained 1.8% and 0.8% statistical uncertainties for the Fe 847-keV and Cr 1434-keV γ rays, respectively. The sample thicknesses were 388 mg/cm^2 for Cr and 164 mg/cm^2 for Fe.

The results are presented in Table 2 where we give both the measured ^{nat}Fe(n,n'γ) cross section including the contributions of the ^{57}Fe(n,2n) reaction, and the ^{56}Fe(n,n'γ) isotopic cross section where we have subtracted the ^{57}Fe(n,2n) contribution of 22 mb using the ENDF/B-VI value at 14.5 MeV corrected for the terrestrial natural abundance of ^{57}Fe of 2.119%.

TABLE 2. Cross sections for the Fe 847-keV γ ray at E_n = 14.5 MeV.

Data Set	σ(^{nat}Fe) (mb) ^{56}Fe(n,n'γ)+^{57}Fe(n,2nγ)	σ(^{nat}Fe) (mb) ^{56}Fe(n,n'γ) natural	σ(^{56}Fe) (mb) ^{56}Fe(n,n'γ) isotopic
This Work - Absolute	705 ± 56	683 ± 57	744 ± 62
This Work - Relative	684 ± 45	669 ± 46	730 ± 50
Simakov *et al.* Evaluation	785 ± 48		
Savin *et al.* Evaluation	621 ± 62		
ENDF/B-VI Inelastic Channel			681

INELASTIC SCATTERING CHANNEL CROSS SECTIONS

For ^{56}Fe, a very large fraction of the inelastic decay cascades down through the first excited state. From this we assume that our value for the ^{56}Fe(n,n'γ) cross section at 14.5 MeV provides a lower limit on the ^{56}Fe inelastic channel cross section that is very close to the true value. In Table 2 we compare our values with those from the ENDF evaluated data library. We note that the ENDF/B-VI cross section is 7% lower than our value.

The evaluations are constrained so that the sum of the various channel cross sections (elastic, inelastic, n-p, n2n, etc) must equal the accurately (< 2%) measured neutron total cross section. If we increase the inelastic cross section by 7% then we must decrease the elastic channel cross section by 7% of the inelastic cross section to maintain the correct total. In Fig. 2 we show the ENDF/B-VI elastic scattering cross section for ^{56}Fe with the available measurements. The dashed line shows the ENDF elastic cross section reduced as discussed above. The improved agreement between the evaluation and the elastic scattering data support a revision of these cross sections.

INTEGRAL MEASUREMENTS AND EVALUATED DATA LIBRARIES

Integral neutron spectral measurements with pulsed spheres and for nuclear reactor pressure vessels, in the past, have often shown more high-energy neutrons than calculated using the data libraries, and hence appear to contradict our result of a larger inelastic cross section. A solution to this problem may lie in adjusting the angular distributions of the neutron elastic and inelastic scattering. New higher accuracy

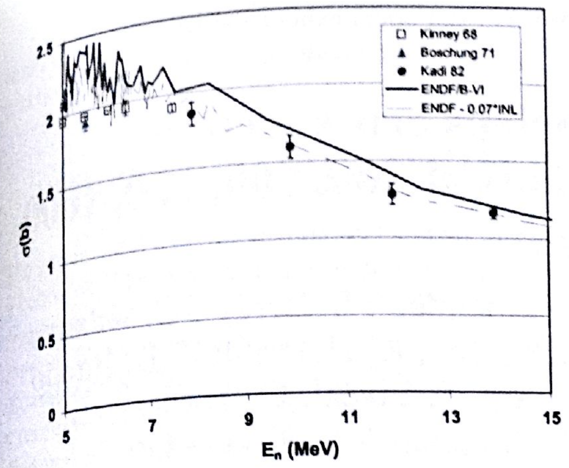

FIGURE 2. Cross-section measurements for ^{56}Fe(n,n) elastic scattering are shown with the current ENDF/B-VI elastic scattering cross section (upper curve) and with the ENDF values reduced by 7% of the inelastic channel cross section (lower curve).

pulsed-sphere data such as those of Massey *et al.* [11] may aid in resolving these questions. Recent measurements of Christodoulou *et al.* [12] of the angular distributions of the elastic and inelastic scattering of neutrons from Fe do show some discrepancies. It is noted that at the very forward angles the experiment underestimates the elastic cross section due to the variation with angle of the incident neutron energy.

CONCLUSION

The data presented here give a more accurate value for production of the 847-keV γ ray, and a consistent set of results for neutron inelastic, elastic, and total reaction cross section values for ^{56}Fe. We plan to improve the accuracy of the measured thickness of our Cr sample and of the relative efficiency measurements to further refine our cross section result. Additional work on interpreting integral measurements on Fe and the evaluated data libraries appears to be needed.

Only the recently updated JEFF 3.0 evaluated library is in good agreement with the present results at $E_n = 14.5$ MeV, although this is near a local maximum. The ENDF/B-VI, JENDL 3.3, and BROND 2 library values are all lower, the first two by about 7% and the latter by 17%.

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